Strong far field coherent scattering of ultraviolet radiation by holococcolithophores.

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By considering the structure of holococcoliths (calcite plates that cover holococcolithophores, a haploid phase of the coccolithophore life cycle) as a photonic structure, we apply a discrete dipolar approximation to study the light backscattering properties of these algae. We show that some holococcolith structures have the ability to scatter the ultraviolet (UV) radiation. This property may represent an advantage for holococcolithophores possessing it, by allowing them to live higher in the water column than other coccolithophores.

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I. INTRODUCTION

Recently, attention is paid to photonic structures in biology as living physical systems generated by evolution (Ref. [1] and references therein). Coccolithophores are unicellular algae distinguished by a covering of calcium carbonate plates, called coccoliths, of still unknown purpose in spite of numerous hypotheses about their possible functions [2, 3, 4]. Coccolithophores are found throughout the euphotic zone of the sea and constitute a significant fraction of the phytoplankton in open ocean environments. They impact greatly on marine ecosystems, and play an important role in the global carbon cycle [5].The optical effects of coccolithophores have been widely investigated [6], but their exact optical properties have not yet been determined. It is known that coccoliths do not absorb light but rather reflect or scatter it by acting as tiny mirrors dispersed in the water [7]; consequently, the main optical impact of coccolithophores is an increase in light scattering. The light backscattering properties of oceanic mesoscale coccolithophore blooms have been measured experimentally. It has also been observed that coccolithophore blooms cause a large increase of the ocean albedo at all light wavelengths but it is particularly outstanding in the threshold of the UV spectrum [7, 8]. The Mie model has been used to fit the experimental measurements, under the assumption that the coccolithophore cell covering (coccosphere) could be represented as a homogeneous sphere [9].

In the present work, we turn our attention to holococcoliths, special coccoliths that have a periodic structure of calcite crystallites and form the cell covering of holococcolithophores (the haploid phase of many coccolithophores). We present a dipolar multiscattering model that considers an ensemble of dielectric calcite nanospheres, arranged according to the structure of some holococcoliths. A dipolar multiscattering model is suitable after considering the size and shape of the coccolith microstructure and the involved frequency range. Neither the diffraction theory can be successfully applied at this dimensions nor the classical optics on calcitic microlenses [10]. In particular, as shown in Figure 1, we will consider holococcoliths that present a triangular layer on top of an hexagonal one of tiny similar-sized calcite crystalline nanospheres (the so-called crystallites). As we shall show, these crystalline structures enhance the light scattering for UV radiation and thus constitute natural calcitic photonic structures.

In the top layers of aquatic environments, phytoplankton receive solar energy, necessary to drive photosynthesis, but are simultaneously exposed to UV radiation that can affect biological processes and damage DNA and other cell compounds [11, 12, 13, 14]. A widespread phytoplankton response is the production of sunscreening compounds such as mycosporine-like amino acids [15]. With regard to coccolithophores, it has been suggested that coccoliths could exert a protective effect by reflecting UV light [3]. According to our results, the crystalline photonic structure of certain holococcoliths enhances UV backscattering. This could have some ecological advantages and could represent an evolutive adaptation of some holococcolithophores, implying a particular choice of the crystalline structure parameters, *i.e.* cell parameter and calcite nanosphere radius, of the holococcoliths.

II. THE MODEL

In Figure 1 images of holococcoliths from *Calcidiscus leptoporus* HOL (formerly *Crystallolithus rigidus*) and *Helicosphaera carteri* HOL, formerly *Syracolithus catilliferus* are shown. Inspired in the observed hexagonal layer of calcite crystals with a triangular layer on top, we

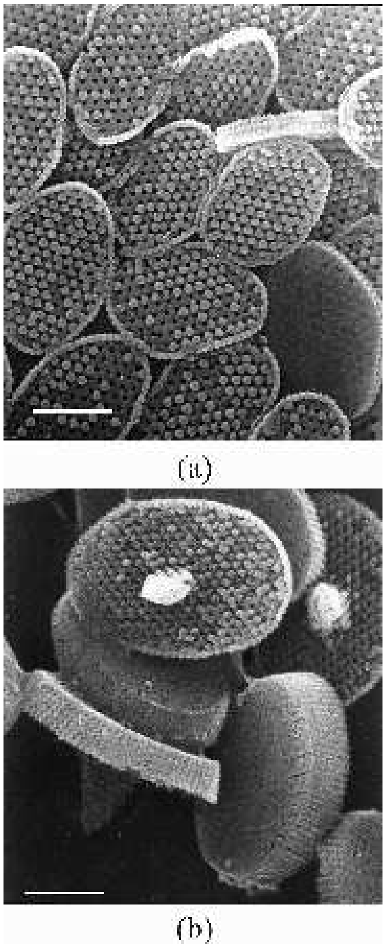

FIG. 1: (a) *Calcidiscus leptoporus* HOL (formerly *Crystallolithus rigidus*). (b) *Helicosphaera carteri* HOL (formerly *Syracolithus catilliferus*). In both cases, images are taken from Ref. [16] (Figs. 62D and 10D, respectively) and the scale bar is 1 μm.

consider the model, shown in Figure 2, that consists of calcium carbonate spheres with a radius of 0.045 μm, arranged in a plane of hexagons with an edge-length of 0.09 μm, covered by a triangular layer with an edge-length of 0.16 μm (triangular and hexagonal layers are 0.09 μm apart). All this structure is immersed in water with a refractive index of 1.333 at a wavelength of 633 nm. Our model assumes a holococcolith surface of 2 $(\mu\text{m})^2$.

Based on the size and shape of the holococcolith microstructure, we adopt a discrete multipolar approximation to study radiation scattering. In this approximation, the target is replaced by an array of point dipoles or, in general, multipoles, which become electromagnetic scatterers. In each vertex, the polarization of the incident radiation field induces an electromagnetic multipole that oscillates with a specific phase, defined by its position in space, and radiates energy in all directions. Far away from the multipole, the angular distribution of the radiation scattered by this structure is given by the coherent

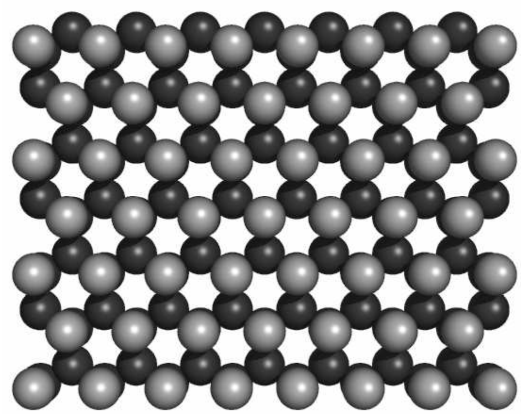

FIG. 2: Model with spheres representing the two layers observed in holococcoliths as those shown in Figure 1. Spheres with a radius of 0.045 μm are arranged in a plane of hexagons (dark spheres) with an edge-length of 0.09 μm, covered by a triangular layer (light gray spheres) with an edge-length of 0.16 μm. The separation between planes is 0.09 μm.

superposition of the emission of each individual source, measured in the test point in space. Thus, in our model, the calcium carbonate spheres arranged in the hexagonal and triangular layers are replaced by point dipoles.

The power radiated in the direction $\mathbf{n}$, with polarization $\boldsymbol{\epsilon}$, per unit of solid angle, that results from the incident radiation in the direction $\mathbf{n}_0$, with polarization $\boldsymbol{\epsilon}_0$ is the derivative of the scattering cross section σ with respect to the solid angle Ω [17]:

$$\frac{d\sigma}{d\Omega} = \frac{k^4}{\left(4\pi\epsilon_0 E_0\right)^2} \left| \sum_j \left[\boldsymbol{\epsilon}^* \cdot \mathbf{p}_j\right] e^{i\mathbf{q}\cdot\mathbf{r}} \right|^2 , \qquad (1)$$

Where $\mathbf{q} = k(\mathbf{n}_0 - \mathbf{n})$, λ is the wave-length, $k = 2\pi n/\lambda$, E_0 is the external electric field and $\mathbf{p}_j$ are the dipolar moments. The sum extends over all nano-spheres. The calculation was carried out assuming that the modeled arrangement has an arbitrary orientation with respect to the incidence direction with the angles (θ, ϕ) for the standard spherical coordinates. The polarization of the incident electromagnetic field is well defined; both S and P polarization are considered.

III. RESULTS

In Figure 3 the backscattering intensity against radiation wave-length is shown for different azimuthal angle incidence (θ). Negligible differences between P and S polarization are measured. We present here results for S-polarization of the incident radiation.

As can be seen in Figure 3, the geometrical structure in Fig. 2 "sees" short wavelengths better than the large ones. For non-grazing incidences the backscattering is

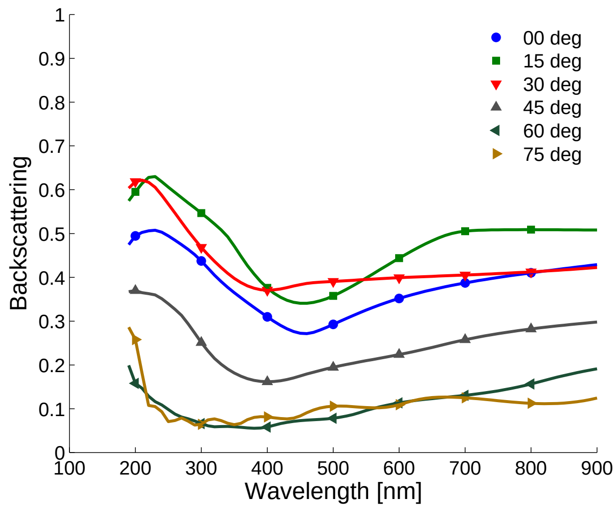

FIG. 3: (Color online) Radiation backscattering of the periodic structure shown in Fig. 2.

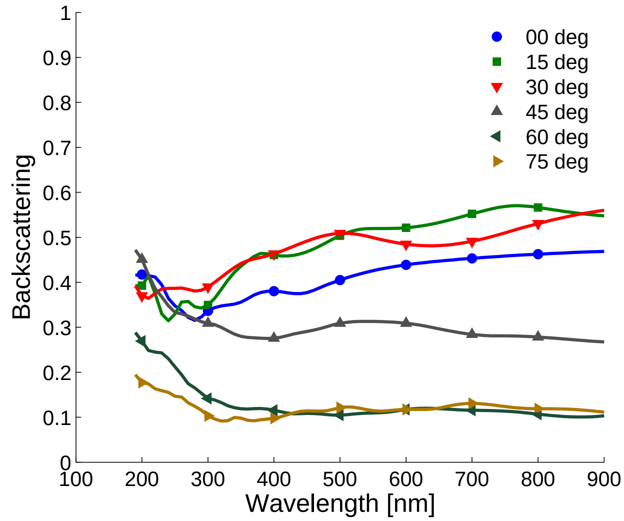

FIG. 4: (Color online) Radiation backscattering of the amorphous structure.

strongly enhanced in the UV range (radiation wavelength less than 400 nm).

With the aim to ascertain the importance of the geometrical arrangement, we compared the optical properties of a hexagonal and an amorphous structure. First, we modeled a geometrical structure using the hexagonal and triangular layers as in Fig. 2. Next, the position of each scatterer was scrambled in a random manner inside a box of $2.54\times1.54\times0.2$ μm. This yields a random (amorphous) structure of dielectric nanospheres (dipoles)

As compared with an amorphous distribution of calcitic spheres, the present case of periodic location for the scatters shows a sharp variation for both wavelength and orientation.

In an amorphous arrangement (Fig. 4), the different plots represent different orientations with respect to the incident electromagnetic field. By adding all the backscattered radiation, we can see an even effect at all wavelengths, with small variation for different orientations. The periodic structure shows a sharp variation for both wavelength and orientation. It also filters more efficiently radiation wavelengths below 400 nm and the total amount of radiation backscattered is clearly greater for this case that for the amorphous one.

IV. DISCUSSION

The arrangement of the calcite crystallites in hexagonal or other geometric patterns is usual in holococcolithophores (ex. *Syracosphaera pulcra* HOL (formerly *Calyptrosphaera oblonga*), *Calyptrolithophora papillifera*, *C. gracillima*, *Holococcolithophora heimdaliae*, *H. dentata*). Among them, a few ones present two or more clearly defined crystallite layers (for example, the here modeled *Calcidiscus leptoporus* HOL and *Helicosphaera carteri* HOL, formerly *Syracolithus catilliferus*). Some other holococcolithophores present a clear hexagonal layer and a careful analysis shows an incipient triangular layer (with an identical arrangement as those shown in Fig. 2) which, we hypothesize, is necessary to scatter the UV light. This is the case, for instance, of *Calyptrolithophora papillifera* and *Syracosphaera pulchra* HOL, formerly *Calyptrosphaera oblonga* HOL [16].

The results produced with the proposed coccolith geometry help to understand published work regarding the interaction between radiation and this complex structure. The simulations for the modeled hexagonal and triangular pattern (Figure 3) reveal that the UV light can be more strongly backscattered than that of other wavelengths. At the same time, the decreased backscattering around 400-700 nm would minimize the loss of photosynthetically active light. It has been shown [18] that holococcolithophores tend to be found higher in the water column than heterococcolithophores, the diploid phase of the coccolithophore life cycle, which have a different coccolith organization. The two-layered coccolith structure of holococcolithophores such as those studied here suggests a possible strategy of increasing the reflection of UV light away from the cell, thus enhancing the ability of the organism to live higher in the water column, as it has been speculated [3] with respect a potential light regulation function of the coccoliths. The differential backscattering of UV light may represent an additional adaptation for the utilization of different ecological niches by cells with diverse coccolith structures.

Acknowledgments

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